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**Abstract**
A potential industrial process for producing a hydrogen-rich synthesis gas by reacting coal char and steam in an electrofluid reactor is described. The characteristics of this type of reactor are reviewed, and a reaction model which appears to fit experimental results is proposed. Product gas compositions and energy requirements predicted by the model for the gasification process are presented for various possible operating conditions. The present state of development of the reaction system and foreseeable problems which must be worked out are reviewed. In addition, the adaptation of the process to the production of various products such as hydrogen, methane, and methanol is discussed.

# Production of Hydrogen from Coal Char in an Electrofluid Reactor

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A potential industrial process for producing a hydrogen-rich synthesis gas by reacting coal char and steam in an electrofluid reactor is described. The characteristics of this type of reactor are reviewed, and a reaction model which appears to fit experimental results is proposed. Product gas compositions and energy requirements predicted by the model for the gasification process are presented for various possible operating conditions. The present state of development of the reaction system and foreseeable problems which must be worked out are reviewed. In addition, the adaptation of the process to the production of various products such as hydrogen, methane, and methanol is discussed.

An investigation of a process for the electrothermal gasification of coal char to produce a hydrogen-rich synthesis gas was initiated several years ago by Iowa State University. In this process, the char is reacted with steam at high temperatures in a fluidized bed reactor which is heated by passing an electric current through the bed of conducting particles. Preliminary results obtained with both 4- and 12-in. diam reactors have been reported (Beeson et al., 1970; Pulsifer et al., 1969). The bench-scale reactor in current use is shown in Figure 1. This reactor has been operated at atmospheric pressure for prolonged periods with both single-phase and three-phase, low-voltage (200 to 400 V) power. Results to date have been most encouraging and together with the results obtained by Kavlick et al. (1970) at high pressures warrant consideration of future large-scale commercial applications. In this regard it may be noted that the Institute of Gas Technology has undertaken to integrate this process with a coal hydrogasification process to produce a methane-rich pipeline gas (Lee, 1970; Schora and Lee, 1969).

Successful industrial applications of electrothermal gasification will depend on careful consideration of the advantages, limitations, and operational characteristics of the process. One of the principal advantages of the process is its versatility. An electrofluid bed reactor can be operated over wide ranges of temperature and pressure, and a wide variety of carbonaceous solids can be employed. By proper choice of operating conditions, it is possible to produce a raw synthesis gas which is relatively rich in hydrogen or alternatively one which is richer in carbon monoxide. It is also possible to select conditions which will provide appreciable concentrations of methane. Electrothermal gasification can be combined with other processes such as shift conversion, methanation, and separation, to produce products ranging from pure hydrogen to various mixtures of hydrogen and carbon monoxide to essentially pure methane. Furthermore, since an electrofluid bed reactor can be operated under high pressures, it is possible to eliminate costly gas compression in many cases. One of the principal limitations of electrothermal gasification is that it utilizes a relatively expensive form of energy. Hence, it can only be applied economically where either low-cost power is available or can be generated. Of course, another limitation is that the means for carrying out the process are not fully developed. However, this should only be a transitory limitation.

To encourage future applications of electrothermal gasification, the principal operation characteristics of this process were developed on the basis of a reaction model which seems to fit the results of bench-scale tests. The operational characteristics include the product gas compositions, yields, and energy requirements for various possible operating conditions. These characteristics are presented below together with a discussion of some possible applications which illustrate how the unique characteristics of the system can be utilized to advantage.

### Reaction Model

Kavlick et al. (1970) listed the following reactions as being the principal ones occurring during the steam gasification of coal char at high temperatures and pressures:

$$C + H_2O = CO + H_2 \quad (1)$$

$$CO + H_2O = CO_2 + H_2 \quad (2)$$

$$C + 2H_2 = CH_4 \quad (3)$$

Reaction 1 is highly endothermic and its completion is favored by a high temperature and a low pressure. Reaction 2 is moderately exothermic and its completion is favored by a low temperature and not at all by pressure. Reaction 3 is highly exothermic and its completion is favored by a low temperature and a high pressure.

The results of Kavlick et al. (1970) indicated that during electrothermal gasification tests conducted at 1000 psig and temperatures between 1700° and 1900°F the last two reactions were usually at equilibrium, whereas the first reaction was the rate-controlling reaction. The results of our work at atmospheric pressure have shown a similar pattern but, of course, at this pressure, the extent of Reaction 3 has generally been very small.

The preceding set of reactions together with certain assumptions provide a useful model for predicting the composition of the gas produced by electrothermal gasification as well as the energy requirements of the process. The chief assumption is that the last two reactions are so fast that they are essentially at equilibrium. Then for a given assumed conversion of

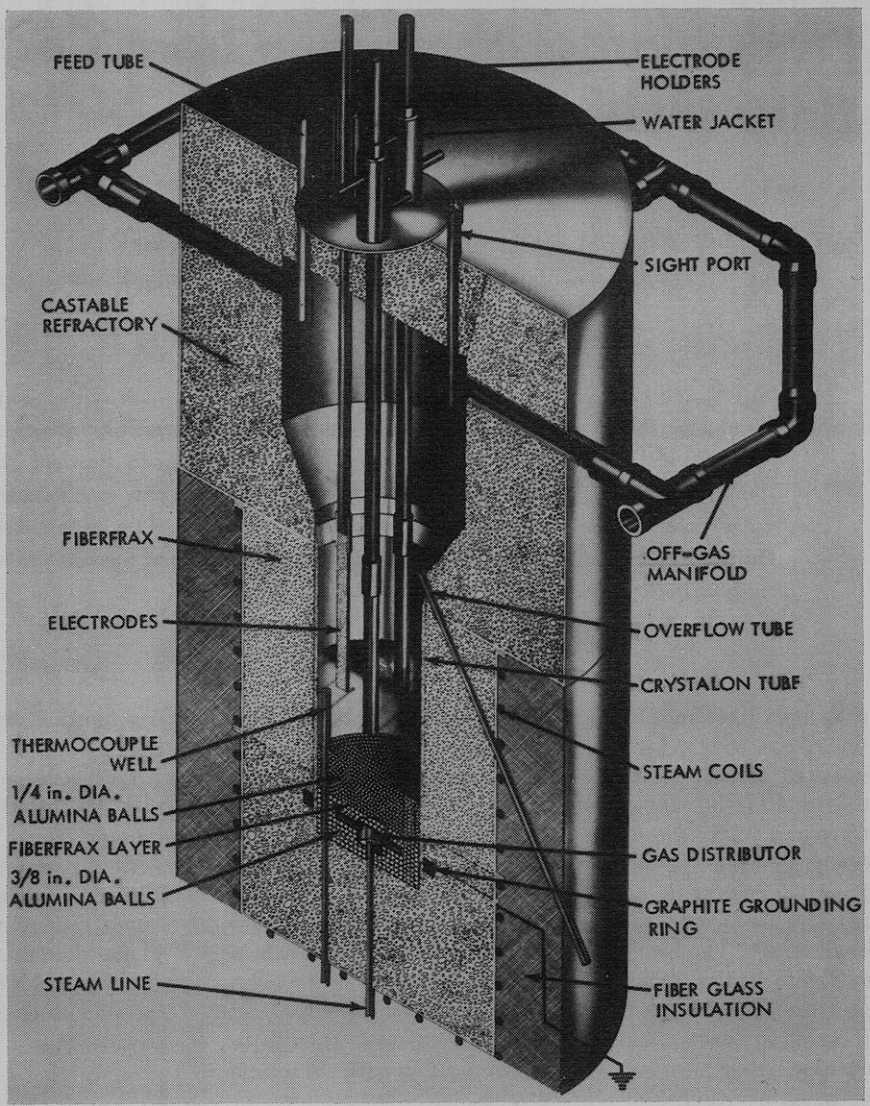


Figure 1. Bench-scale (12-in. i.d.) electro-fluid bed reactor

**Table I. Comparison of Predicted and Observed Gas Compositions**

| Reactor | Temp, °F | Press., psig | Steam velocity, ft/sec | Steam conv., % | Comp. (dry basis), mol % | | |
|---|---|---|---|---|---|---|---|
| | | | | | | Predicted | Observed |
| 4-in. diam (run 11) | 1600 | 0 | 0.14 | 51 | CO | 23 | 20 |
| | | | | | $CO_2$ | 18 | 20 |
| | | | | | $H_2$ | 59 | 58 |
| 4-in. diam (run 17) | 1600 | 0 | 0.14 | 58 | CO | 25 | 26 |
| | | | | | $CO_2$ | 16 | 15 |
| | | | | | $H_2$ | 59 | 56 |
| 4-in. diam (run 19) | 1600 | 0 | 0.41 | 37 | CO | 17 | 24 |
| | | | | | $CO_2$ | 22 | 14 |
| | | | | | $H_2$ | 61 | 58 |
| 12-in. diam (run 27) | 1700 | 0 | 0.05 | 66 | CO | 32 | 38 |
| | | | | | $CO_2$ | 12 | 8 |
| | | | | | $H_2$ | 56 | 54 |
| IGT (run EG-46) | 1900 | 1008 | 0.21 | 65 | CO | 34 | 33 |
| | | | | | $CO_2$ | 13 | 12 |
| | | | | | $H_2$ | 45 | 46 |
| | | | | | $CH_4$ | 8 | 9 |
| IGT (run EG-48) | 1900 | 1010 | 0.24 | 53 | CO | 28 | 27 |
| | | | | | $CO_2$ | 17 | 14 |
| | | | | | $H_2$ | 48 | 51 |
| | | | | | $CH_4$ | 7 | 8 |

steam by the first reaction it is possible to solve appropriate material balance and equilibrium expressions for the final gas composition. Of course, the limiting steam conversion corresponds to the situation where the first reaction is also at equilibrium. Additional assumptions which greatly simplify the calculations are that the gases behave ideally at the reactor temperature and that the activity of carbon is unity. Values of the thermodynamic equilibrium constants are recorded in the literature (von Fredersdorff and Elliott, 1963).

To verify this model, values of the gas composition predicted by the model were compared with gas compositions observed during operation of both our 4- and 12-in. diam electrofluid beds (Beeson et al., 1970; Pulsifer et al., 1969) and with gas compositions observed by Kavlick et al. (1970) in the operation of the high-pressure unit at the Institute of Gas Technology. Some of these results are presented in Table I (where the total steam conversion is listed rather than just that portion converted by Reaction 1). In the case of our 4-in. diam reactor where a fluidized bed depth of 22 in. was employed, the agreement between predicted and observed values was good at lower steam velocities (and consequently higher steam conversions) but not at higher velocities. The agreement was poorer with the 12-in. diam reactor, perhaps due to the use of a shallower fluidized bed depth (12 in.). Obviously the agreement was good in the case of the Institute of Gas Technology (IGT) runs.

### Operational Characteristics

Since the reaction model described above seemed to represent the gasification process fairly well, it was used to predict the operational characteristics of the process. This included a prediction of the gas composition, energy requirements, and yields which would result from gasifying carbon under various selected temperatures (1600°, 1900°, and 2200°F), pressures (1, 70, and 200 atm), and conversions. For this preliminary and rather general analysis it was assumed that the solid reactant would be pure carbon. In a more detailed and exacting analysis, the minor constituents of the solid reactant, such as hydrogen, sulfur, and nitrogen, would need to be considered.

The gas compositions which were predicted to result from gasifying carbon at selected conditions are summarized in Table II. It should be noted that values of the conversions $x_2$ and $x_3$ were also predicted. From this data it is readily apparent that a wide range of gas compositions can be obtained by electrothermal gasification. Several major trends are apparent. As the steam converted by Reaction 1 increases, the steam converted by Reaction 2 decreases, and the carbon converted to methane by Reaction 3 increases. The maximum or equilibrium conversion of steam (by Reaction 1) rises with increasing temperature but falls with increasing pressure. The concentrations of carbon monoxide, hydrogen, and methane all rise with increasing steam conversion, whereas the concentration of carbon dioxide tends to fall. The ratio of hydrogen to carbon monoxide decreases with increasing steam conversion. The concentration of methane rises with increasing pressure but falls with rising temperature.

The overall isothermal heat of reaction calculated by employing the expression

$$\Delta H_r = x_1\Delta H_1 + x_2\Delta H_2 + x_3\Delta H_3 \qquad (4)$$

is also listed in Table II for each combination of chosen conditions. This heat of reaction represents the amount of heat taken up by the gasification process for each mole of steam fed when all components of the reaction system are at the same temperature. Standard heats of reaction were taken from the literature (von Fredersdorff and Elliott, 1963). No allowance was made for the effect of pressure since it was assumed that the gases behave ideally.

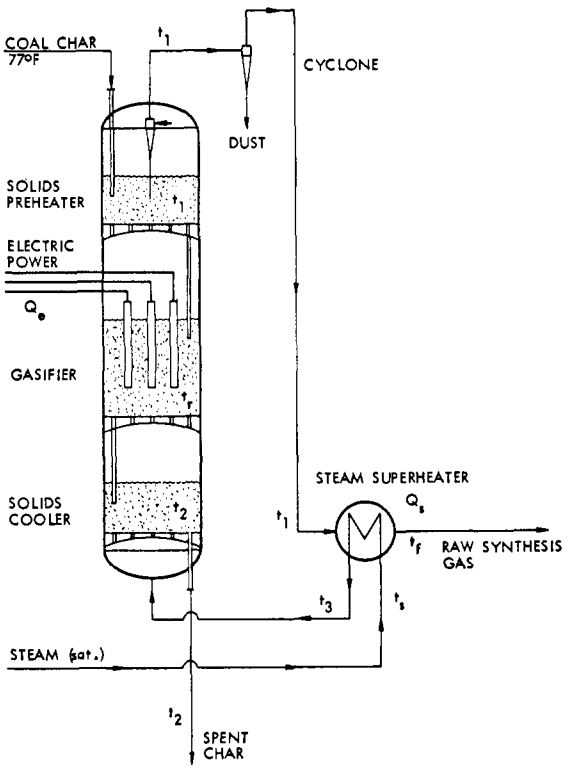


**Figure 2. Electrothermal gasifier and associated equipment for heat recovery**

When evaluating future applications of electrothermal gasification, it is important to consider the total energy required for gasification. The amount of energy needed will not only depend on the conditions chosen for gasification but also on the extent to which heat economy is practiced. Although there are many ways of conserving energy, a fairly typical method is illustrated by Figure 2. In this method the reactants and products exchange heat in a series of countercurrent heat transfer steps. The design utilizes a three-compartment fluidized solids unit. In the uppermost compartment, coal char is preheated by direct contact with the outflowing gases, while in the lowest compartment, the spent char is cooled by contact with the incoming steam. Additional heat is recovered from the outflowing gases by heat exchange with the incoming steam.

For the present analysis, the scheme of Figure 2 was utilized as a basis for estimating the energy requirements of electrothermal gasification. The total energy which would have to be supplied electrically ($Q_e$), the heat transferred in the steam superheater ($Q_s$), and various temperatures ($t_1$, $t_2$, and $t_3$) were estimated by employing a series of energy balances. For this purpose it was assumed that char would be supplied at ambient temperature (77°F) and saturated steam at reactor pressure. It was necessary to specify the temperature of the synthesis gas leaving the steam superheater as well as both the steam and carbon conversions. Pressure drop in the system was neglected. Although the gases in the reactor were assumed ideal, the effect of pressure on the enthalpy of gases in the steam superheater was taken into account where the effect was significant.

**Table II. Predicted Gas Composition and Overall Heat of Reaction**

| $x_1$ | $x_2$ | $x_3$ | Composition, mol fraction $H_2O$ | CO | $H_2$ | $CO_2$ | $CH_4$ | $\Delta H_r$, Btu/mol |
|---|---|---|---|---|---|---|---|---|
| | | | ($p$ = 1 atm and $t$ = 1600°F) | | | | | |
| 0.343 | 0.157 | 0.004 | 0.373 | 0.139 | 0.367 | 0.117 | 0.003 | 17,600 |
| 0.617 | 0.133 | 0.008 | 0.155 | 0.299 | 0.459 | 0.082 | 0.005 | 33,800 |
| 0.829 | 0.071 | 0.011 | 0.055 | 0.417 | 0.483 | 0.039 | 0.006 | 47,000 |
| 0.959[a] | 0.018 | 0.011 | 0.012 | 0.483 | 0.490 | 0.009 | 0.006 | 55,300 |
| | | | ($p$ = 1 atm and $t$ = 1900°F) | | | | | |
| 0.371 | 0.129 | ... | 0.365 | 0.177 | 0.365 | 0.094 | ... | 19,900 |
| 0.652 | 0.098 | ... | 0.151 | 0.335 | 0.454 | 0.059 | ... | 36,700 |
| 0.852 | 0.048 | ... | 0.054 | 0.434 | 0.486 | 0.026 | ... | 49,000 |
| 0.994[a] | 0.002 | ... | 0.002 | 0.497 | 0.499 | 0.001 | ... | 58,000 |
| | | | ($p$ = 1 atm and $t$ = 2200°F) | | | | | |
| 0.391 | 0.109 | ... | 0.359 | 0.203 | 0.359 | 0.078 | ... | 21,300 |
| 0.673 | 0.077 | ... | 0.149 | 0.356 | 0.448 | 0.046 | ... | 38,100 |
| 0.864 | 0.036 | ... | 0.054 | 0.444 | 0.483 | 0.019 | ... | 49,800 |
| 0.999[a] | 0.000 | ... | 0.001 | 0.499 | 0.500 | 0.000 | ... | 58,100 |
| | | | ($p$ = 70 atm and $t$ = 1600°F) | | | | | |
| 0.300 | 0.184 | 0.107 | 0.432 | 0.097 | 0.226 | 0.154 | 0.090 | 10,700 |
| 0.400 | 0.203 | 0.142 | 0.316 | 0.157 | 0.254 | 0.161 | 0.113 | 14,900 |
| 0.500 | 0.202 | 0.171 | 0.224 | 0.224 | 0.271 | 0.152 | 0.129 | 19,600 |
| 0.511[a] | 0.201 | 0.174 | 0.216 | 0.232 | 0.272 | 0.150 | 0.130 | 20,200 |
| | | | ($p$ = 70 atm and $t$ = 1900°F) | | | | | |
| 0.355 | 0.145 | 0.058 | 0.386 | 0.162 | 0.296 | 0.112 | 0.045 | 16,400 |
| 0.454 | 0.146 | 0.076 | 0.290 | 0.224 | 0.325 | 0.106 | 0.055 | 21,500 |
| 0.565 | 0.135 | 0.096 | 0.204 | 0.293 | 0.346 | 0.092 | 0.065 | 27,300 |
| 0.772[a] | 0.085 | 0.119 | 0.087 | 0.416 | 0.374 | 0.051 | 0.072 | 39,200 |
| | | | ($p$ = 70 atm and $t$ = 2200°F) | | | | | |
| 0.383 | 0.117 | 0.029 | 0.369 | 0.196 | 0.326 | 0.086 | 0.021 | 19,600 |
| 0.603 | 0.097 | 0.047 | 0.193 | 0.325 | 0.389 | 0.062 | 0.030 | 31,900 |
| 0.859 | 0.041 | 0.066 | 0.056 | 0.456 | 0.428 | 0.023 | 0.037 | 46,800 |
| 0.924[a] | 0.023 | 0.071 | 0.029 | 0.486 | 0.434 | 0.012 | 0.038 | 53,500 |
| | | | ($p$ = 200 atm and $t$ = 1600°F) | | | | | |
| 0.300 | 0.206 | 0.158 | 0.433 | 0.082 | 0.166 | 0.181 | 0.138 | 8,400 |
| 0.400 | 0.231 | 0.205 | 0.309 | 0.141 | 0.185 | 0.193 | 0.172 | 12,000 |
| 0.421[a] | 0.234 | 0.214 | 0.286 | 0.155 | 0.188 | 0.194 | 0.177 | 12,900 |
| | | | ($p$ = 200 atm and $t$ = 1900°F) | | | | | |
| 0.300 | 0.155 | 0.091 | 0.451 | 0.120 | 0.226 | 0.128 | 0.075 | 11,800 |
| 0.400 | 0.166 | 0.121 | 0.340 | 0.183 | 0.253 | 0.130 | 0.095 | 16,300 |
| 0.500 | 0.164 | 0.148 | 0.248 | 0.248 | 0.272 | 0.121 | 0.109 | 21,100 |
| 0.600 | 0.149 | 0.171 | 0.176 | 0.316 | 0.285 | 0.104 | 0.120 | 26,200 |
| 0.635[a] | 0.140 | 0.178 | 0.155 | 0.340 | 0.288 | 0.096 | 0.122 | 28,100 |
| | | | ($p$ = 200 atm and $t$ = 2200°F) | | | | | |
| 0.300 | 0.123 | 0.048 | 0.461 | 0.141 | 0.260 | 0.098 | 0.039 | 13,900 |
| 0.400 | 0.128 | 0.067 | 0.354 | 0.204 | 0.296 | 0.096 | 0.050 | 18,900 |
| 0.500 | 0.122 | 0.084 | 0.267 | 0.267 | 0.321 | 0.086 | 0.059 | 24,200 |
| 0.600 | 0.109 | 0.100 | 0.194 | 0.327 | 0.340 | 0.073 | 0.066 | 29,500 |
| 0.700 | 0.089 | 0.114 | 0.133 | 0.385 | 0.354 | 0.056 | 0.072 | 35,000 |
| 0.800 | 0.064 | 0.127 | 0.081 | 0.440 | 0.365 | 0.038 | 0.076 | 40,700 |
| 0.834[a] | 0.054 | 0.132 | 0.066 | 0.458 | 0.367 | 0.032 | 0.077 | 42,600 |

[a] Equilibrium value.

The estimated temperatures and energy requirements for the process illustrated by Figure 2 are given in Table III for representative operating conditions. In this table, the required electrical energy ($Q_e$) and heat transferred in the steam superheater ($Q_s$) are based on the carbon fed to the reactor. Another quantity given in Table III is the steam-to-carbon feed ratio ($F$) which was calculated from the given conversions by using the relation

$$F = X/(x_1 + x_3) \tag{5}$$

Since synthesis gas ($H_2$ + CO) is the primary product of electrothermal gasification, the yields and energy requirements for producing it are of major importance. The theoretical yields were determined by employing the expressions

$$\text{Yield on carbon} = \frac{2X(x_1 - x_3)}{x_1 + x_3} \tag{6}$$

**Table III. Energy Requirements for Process Shown in Figure 2**

| $P$, atm | $t_r$, °F | $X$ | $x_1$ | $F$ | $t_s$, °F | $t_f$, °F | $t_1$, °F | $t_2$, °F | $t_3$, °F | $Q_s$, Btu/mol | $Q_e$, Btu/mol |
|---|---|---|---|---|---|---|---|---|---|---|---|
| 1 | 1600 | 0.9 | 0.959[a] | 0.928 | 212 | 700 | 1263 | 1163 | 1137 | 7,600 | 57,100 |
| 1 | 1900 | 0.9 | 0.994[a] | 0.905 | 212 | 800 | 1483 | 1374 | 1342 | 9,200 | 59,600 |
| 1 | 2200 | 0.9 | 0.999[a] | 0.901 | 212 | 800 | 1708 | 1719 | 1691 | 12,300 | 59,700 |
| 70 | 1600 | 0.9 | 0.511[a] | 1.313 | 548 | 700 | 1323 | 1261 | 1248 | 10,700 | 32,500 |
| 70 | 1900 | 0.9 | 0.772[a] | 1.010 | 548 | 800 | 1507 | 1457 | 1434 | 10,200 | 46,300 |
| 70 | 1900 | 0.9 | 0.565 | 1.362 | 548 | 800 | 1571 | 1472 | 1457 | 14,000 | 45,000 |
| 70 | 1900 | 0.9 | 0.454 | 1.698 | 548 | 800 | 1613 | 1483 | 1470 | 17,700 | 45,400 |
| 70 | 1900 | 0.9 | 0.355 | 2.179 | 548 | 800 | 1658 | 1497 | 1488 | 23,100 | 46,200 |
| 70 | 2200 | 0.9 | 0.924[a] | 0.905 | 548 | 950 | 1709 | 1679 | 1647 | 11,100 | 53,600 |
| 200 | 1600 | 0.9 | 0.421[a] | 1.417 | 692 | 800 | 1340 | 1027 | 1010 | 10,700 | 28,700 |
| 200 | 1600 | 0.9 | 0.300 | 1.965 | 692 | 800 | 1392 | 1064 | 1053 | 15,800 | 29,400 |
| 200 | 1900 | 0.9 | 0.635[a] | 1.107 | 692 | 800 | 1528 | 1287 | 1261 | 11,600 | 40,100 |
| 200 | 2200 | 0.9 | 0.834[a] | 0.932 | 692 | 800 | 1717 | 1582 | 1548 | 12,700 | 48,500 |
| 200 | 1600 | 0.5 | 0.421[a] | 0.787 | 692 | 800 | 1198 | 1009 | 872 | 17,500 | 33,900 |
| 200 | 1900 | 0.5 | 0.635[a] | 0.615 | 692 | 800 | 1335 | 1258 | 1018 | 4,700 | 24,300 |
| 200 | 2200 | 0.5 | 0.834[a] | 0.518 | 692 | 800 | 1478 | 1540 | 1220 | 5,200 | 30,700 |

[a] Equilibrium value.

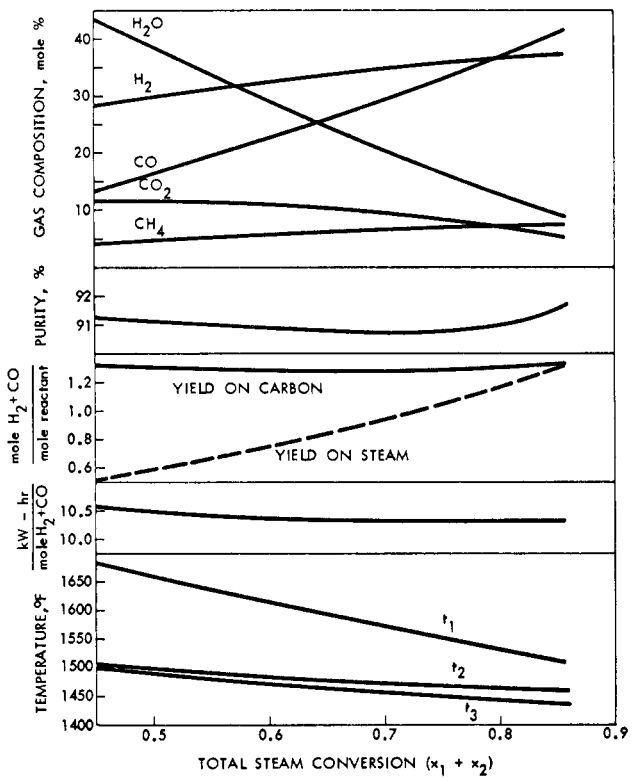


Figure 3. Effect of steam conversion on gasification at 70 atm and 1900°F

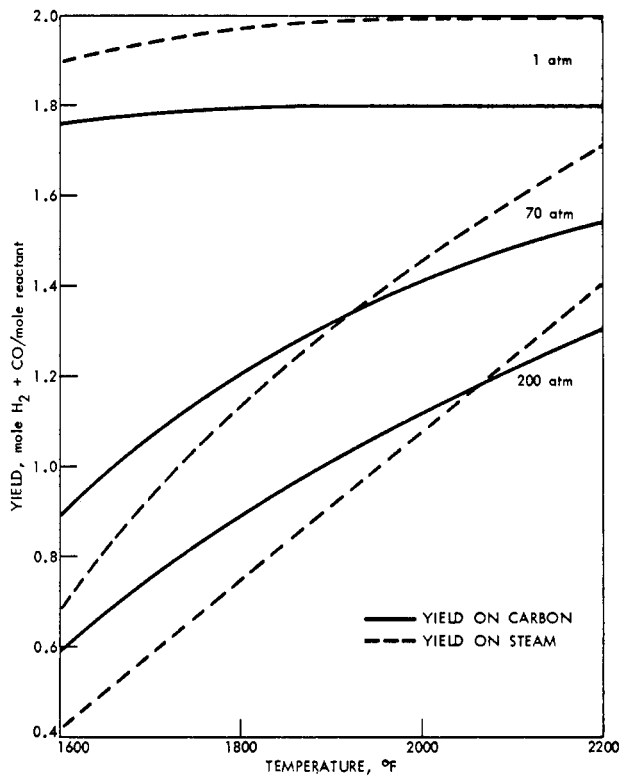


Figure 4. Yields of synthesis gas obtained with equilibrium steam conversion and 90% carbon conversion

$$\text{Yield on steam} = 2\,(x_1 - x_3) \tag{7}$$

and the kilowatt hours of electrical energy needed to produce each mole of synthesis gas were estimated by using the relation

$$\text{Electrical energy} = \frac{2.93(10^{-4})Q_e}{\text{Yield on C}} \tag{8}$$

Another item of importance is the product purity which can be obtained economically. Since relatively economical methods are available for removing water vapor and carbon dioxide, it was assumed that methane would be the only contaminant of the purified synthesis gas. On this basis, the ultimate purity is given by

$$\%\ \text{purity} = \left(\frac{x_1 - x_3}{x_1 - 0.5\,x_3}\right)100 \tag{9}$$

The estimated yields, energy requirements, and ultimate purity of synthesis gas are presented in Figures 3–6 for representative operating conditions. In Figure 3 these quantities are shown as a function of the total steam conversion (conversions ranging up to and including the equilib-

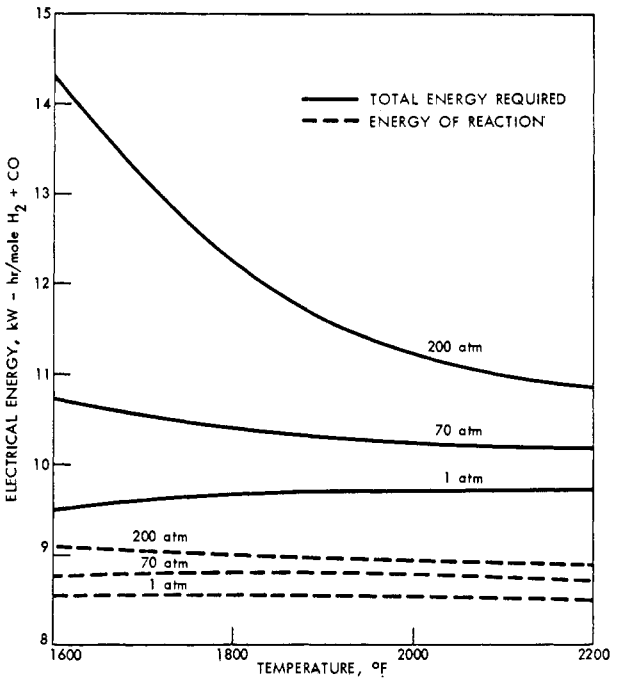


Figure 5. Energy required for gasification based on 90% carbon conversion and equilibrium steam conversions

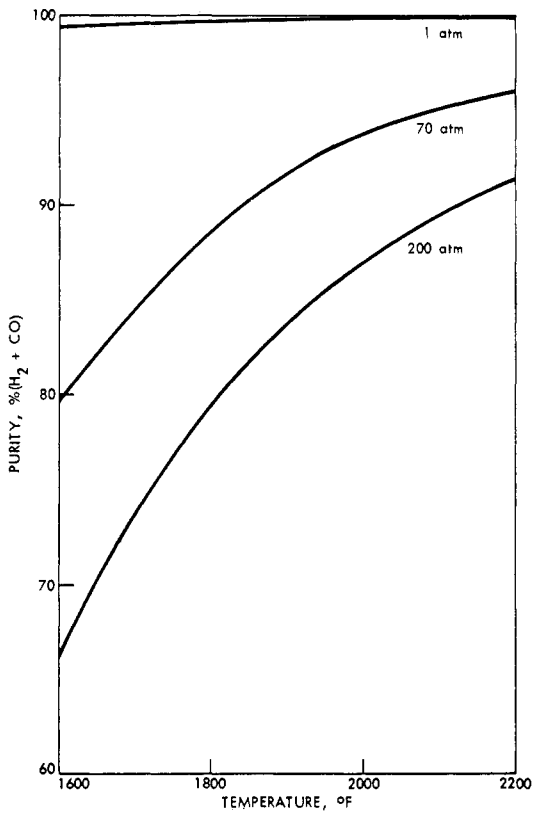


Figure 6. Purity of synthesis gas freed of all contaminants except methane

rium value) for gasification at 70 atm and 1900°F and with a carbon conversion of 90%. Also shown are important system temperatures (identified in Figure 2) and the composition of the reactor off-gas. Interestingly enough, the ultimate purity, yield on carbon, and required input of electrical energy do not vary greatly with steam conversion. However, at the equilibrium steam conversion, the ultimate purity and yield on carbon are a maximum, and the energy requirement is a minimum. As might be expected, the yield based on steam fed decreases markedly as the steam conversion is reduced. On the other hand, the ratio of hydrogen to carbon monoxide increases from 0.9 to 2.1 as the steam conversion is reduced over the indicated range.

Since the major operational characteristics are not strongly influenced by steam conversion, they are presented in Figures 4–6 for gasification under equilibrium conditions only. In Figure 4, it can be seen that the yields of synthesis gas are greatly affected by temperature and pressure. Both the yields on carbon and on steam rise with increases in temperature but fall with increases in pressure. However, at 1 atm the effect of temperature is small. Maximum yields can be obtained by operating at the highest temperature and lowest pressure.

The influence of temperature and pressure on the quantity of energy required to produce a given amount of synthesis gas is illustrated by Figure 5. At the lower pressures the effect of temperature is small, but at the highest pressure it is large. Thus at 200 atm the total energy required decreases markedly when the temperature is raised. The effect of pressure is most pronounced at the lowest temperature and is least pronounced at the highest temperature. At all temperatures, though, an increase in pressure produces an increase in the energy requirement. Gasification at the lowest temperature and pressure would require the least amount of energy.

In addition to the total energy required, the overall reaction energy for operation at various temperatures and pressures is also shown in Figure 5. This was determined by employing the relation

$$\text{Energy of reaction} = \frac{2.93(10^{-4})F\Delta H_r}{\text{Yield on C}} \qquad (10)$$

The difference between the total energy required and the energy of reaction is simply the energy absorbed in heating the reactants to the temperature of the gasifier. Obviously this difference is large, and a more efficient method of preheating the reactants than that portrayed by Figure 2 could reduce the total electrical energy requirement significantly.

Figure 6 shows that the gasifier temperature and pressure can have an important effect on the ultimate purity of the synthesis gas after it is freed of water vapor and carbon dioxide. Only small amounts of methane are produced at 1 atm and, hence, a very pure product can be produced at this pressure. At high pressures, appreciable quantities of methane are produced and the purity of the synthesis gas suffers. However, the purity can be improved by increasing the temperature to the highest possible value.

The effect of operating conditions on gasifier diameter is interesting (Figure 7). By starting with the relation

$$\text{Cross-sectional area} = \frac{\text{volumetric flow rate}}{\text{fluidizing gas velocity}} \qquad (11)$$

and fixing the fluidizing gas velocity, carbon feed rate, and conversion, it can be shown that the reactor diameter is proportional to the quantity shown below:

$$\text{Diameter} \propto \left[\frac{(1 + x_1 - x_3)}{(x_1 + x_3)}\frac{T}{p}\right]^{1/2} \qquad (12)$$

The quantity in brackets approaches a minimum value (4.43) for an equilibrium steam conversion ($x_1$ = 0.421) at

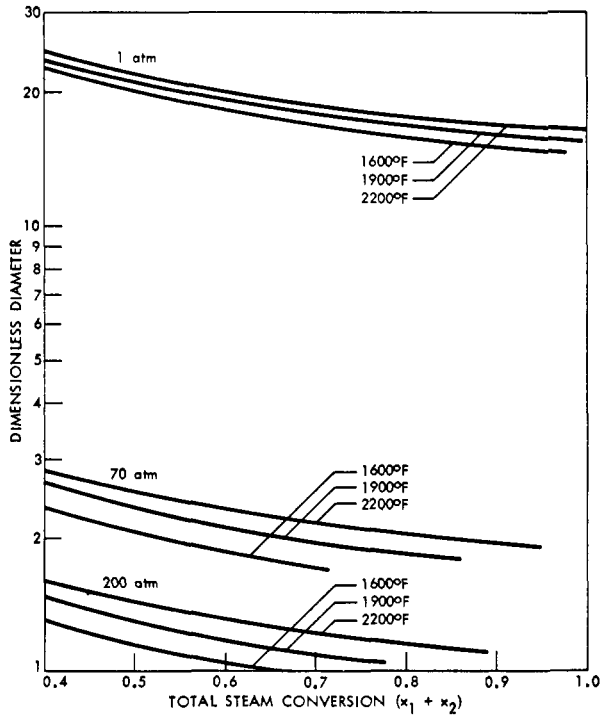


Figure 7. Effect of operating conditions on gasifier size for fixed carbon feed rate and conversion

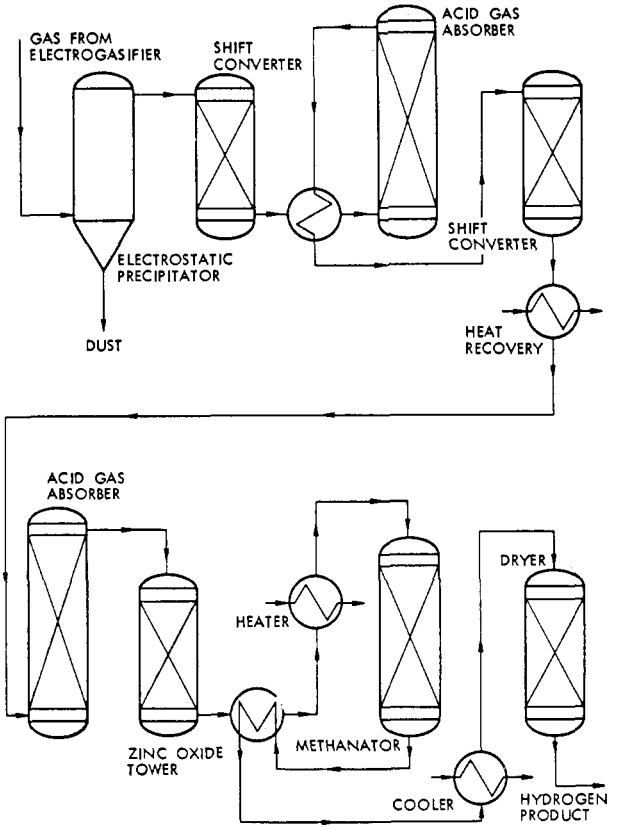


Figure 8. Gas purification, shift conversion, and methanation steps required for the recovery of hydrogen

the highest selected pressure (200 atm) and lowest selected temperature (1600°F). This provides a convenient basis for a dimensionless reactor diameter. Thus the dimensionless diameter determined by the expression

$$D = \frac{1}{4.43}\left[\frac{(1 + x_1 - x_3)}{(x_1 + x_3)}\frac{T}{p}\right]^{1/2} \qquad (13)$$

is simply the ratio of the required diameter at any selected set of conditions to the diameter at the reference set of conditions. Figure 7 is based on the preceding expression and illustrates graphically how the diameter is affected by choice of operating conditions.

### Some Possible Applications

There appear to be a number of potential industrial applications for electrothermal gasification since hydrogen and/or carbon monoxide can be utilized in many different ways. Some of the possibilities are described below along with a discussion of the factors which have an important bearing on process efficiency and economics.

**Hydrogen Production.** Electrothermal gasification can be readily adapted to the efficient production of hydrogen by proper selection of operating conditions and by the addition of appropriate gas purification and carbon monoxide shift conversion steps. Figures 3 to 7 can be used in the selection of conditions for gasification. These diagrams can be applied directly for this purpose since carbon monoxide and hydrogen are essentially equivalent due to the ease with which carbon monoxide can be shift-converted by Reaction 2 over a bed of catalyst. Of course, the anticipated yields and purity of hydrogen would be slightly less than the values shown in these diagrams because it is not economically feasible to convert all of the carbon monoxide. (Typically the gas from a two-stage shift conversion operation will contain from 0.25 to 0.50% carbon monoxide.) For the same reason, the electrical energy required per mole of hydrogen would be slightly greater than that shown.

In many cases hydrogen is required for subsequent operations which are carried out at high pressure. By generating hydrogen at high pressure, compression costs can be eliminated. However, reference to Figures 3–6 shows that some sacrifice must be made in yields, product purity, and energy utilization. Only a careful evaluation of the economics and product requirements would reveal the best choice of operating conditions. Nevertheless, some trends are apparent. Thus if high-pressure gasification is employed, product yields and purity and energy utilization would be maximized by employing the highest possible temperature. Other considerations such as electrode life or the ash fusion temperature would determine the upper temperature limit. On the other hand, if low-pressure gasification is used, the choice of temperature is not very critical as far as yields, product purity, and energy utilization are concerned. For this case, the effect of temperature on the rate of reaction may be the most important factor.

Several steps may be required to recover a usable product from the gasifier off-gas. Although many variants are possible, the processing scheme illustrated by Figure 8 would be fairly typical. In this scheme, dust is removed by a hot electrostatic precipitator, and part of the carbon monoxide is converted to hydrogen and carbon dioxide in the first shift converter. The converter employs a standard high-temperature, chromium-promoted iron catalyst which Wilson and Plants (1968) have found to be sulfur-resistant. The gas is cooled, scrubbed to remove the bulk of the carbon dioxide, and reheated before entering the second shift converter. Most of the remaining carbon monoxide is converted in the

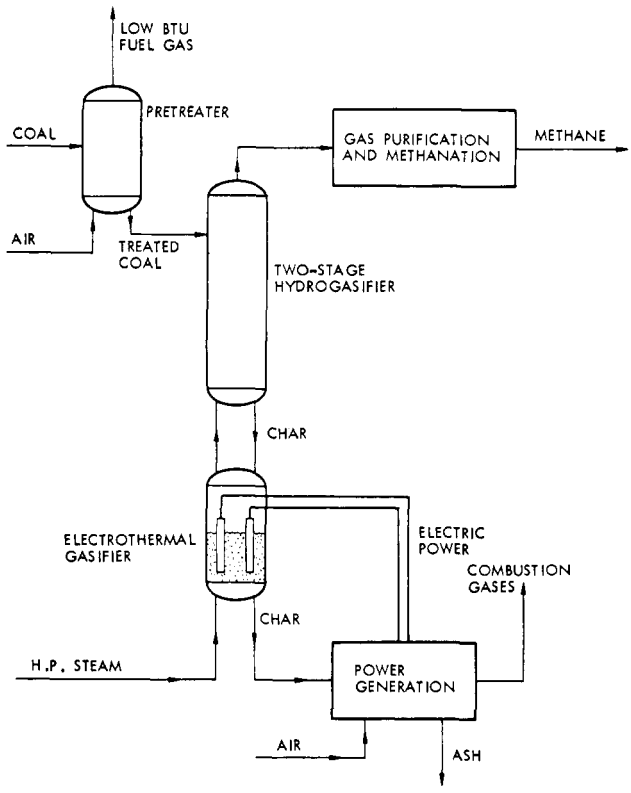


**Figure 9. The Hygas process being developed by the Institute of Gas Technology**

second converter. The gas is then cooled and scrubbed to remove additional carbon dioxide. Although the greater part of any hydrogen sulfide produced during gasification would be removed by the carbon dioxide scrubbers, a tower packed with zinc or iron oxide is provided to remove final traces since the catalyst used in the next step is easily poisoned. After passing through the zinc oxide tower, the gas is reheated and passed over a catalyst which converts residual carbon oxides to methane. The gas is then cooled and dried. The final product is essentially free of all contaminants except methane and any nitrogen which may have been present in the coal char. If these contaminants cannot be tolerated, they can be removed by low temperature adsorption on activated carbon or in other ways.

Obviously the cost of recovering hydrogen depends on the purity required. If small amounts of carbon oxides can be tolerated, the methanation step can be eliminated, and if larger amounts can be tolerated, the second shift converter and carbon dioxide scrubber can also be removed.

**Production of $H_2$–CO Mixtures.** A wide range of hydrogen–carbon monoxide mixtures can be produced by electrothermal gasification (Table II, Figure 3). By the addition of steps for gas purification and, in some cases, shift conversion, it is possible to produce gas suitable for the synthesis of various products. Three cases will be mentioned here: the preparation of synthesis gas for the oxo process, methanol synthesis, and hydrogasification of coal.

The conventional oxo process for the production of aldehydes and alcohols requires a synthesis gas which has a hydrogen-to-carbon monoxide ratio of from 1.0 to 1.24 (Roelen and Beery, 1952; van den Berg, 1970). Such a gas mixture can be readily produced by electrothermal gasification over a wide range of temperature and pressure. Hence, only gas purification steps should be required to recover oxo synthesis gas from the gasifier off-gas. The gas purification system would have to include equipment for dust removal, carbon dioxide absorption, and sulfur removal. Since the oxo process is carried out at pressures in the range of 200 to 300 atm (Roelen and Beery, 1952; van den Berg, 1970), consideration should be given to gasification at high pressure to reduce or eliminate the cost of gas compression. The main problem, of course, would be the formation of methane which is relatively difficult to remove. Reference can be made to Figures 3–6 to estimate yields, purity, and energy requirements for producing oxo synthesis gas.

The synthesis gas used for producing methanol is typically a mixture of hydrogen, carbon monoxide, and carbon dioxide (Hedley et al., 1970). Hydrogen should be present in sufficient concentration to convert the carbon oxides to methanol in accordance with the equations

$$CO + 2H_2 = CH_3OH \quad (14)$$

$$CO_2 + 3H_2 = CH_3OH + H_2O \quad (15)$$

A review of the gas compositions listed in Table II show that the ratio of hydrogen to carbon oxides is generally too low for use directly in methanol synthesis. The ratio can be raised to a suitable level either by removing carbon dioxide or by shifting part of the carbon monoxide to hydrogen and carbon dioxide and scrubbing out the latter. The gas must be desulfurized to prevent poisoning of the methanol synthesis catalyst. Since methanol is produced at pressures ranging from 50 to 300 atm (Hedley et al., 1970) consideration should be given to gasification at high pressure to avoid gas compression. Gasification at 50 atm and 2200°F should yield a methanol synthesis gas with no more than 3% methane. Gasification at higher pressures would, of course, lead to greater concentrations of methane which would present more of a problem. Yields, purity, and energy requirements for methanol synthesis gas can be estimated by referring to Figures 3–6.

The coal hydrogasification process being developed by the Institute of Gas Technology to manufacture a methane-rich substitute for natural gas can directly utilize the raw synthesis gas produced by electrothermal gasification (Lee, 1970; Schora and Lee, 1969). Figure 9 illustrates the proposed manner for integrating electrothermal gasification with hydrogasification. In the integrated process, the electrothermal gasifier operates at essentially the same pressure (about 7 atm) as the hydrogasifier but at somewhat higher temperature (1800–1900°F as opposed to 1700–1800°F in the second stage of the hydrogasifier). The electrothermal gasifier receives hot char from the hydrogasifier and converts a portion of it into synthesis gas which is returned to the hydrogasifier. The remainder of the char is utilized for generating the electric power consumed by the gasifier. The overall thermal efficiency of the integrated process is high, and the estimated energy consumption in the gasifier based on a bituminous coal feed is 12.4 kW-hr/mol $CH_4$ (Knabel and Tsaros, 1967; Tsaros et al., 1969). The economics of producing synthesis gas by this route appear attractive in comparison with other alternatives (Schora and Lee, 1969; Knabel and Tsaros, 1967; Tsaros et al., 1969).

**Methane Production.** Where production of methane from char rather than coal is desired, a somewhat different process than the one described above could be used. In the alternate process, the electrothermal gasifier would be operated at a lower temperature so that an appreciable amount of methane would be produced in the gasifier. Hydrogasification would be bypassed, and the raw synthesis gas

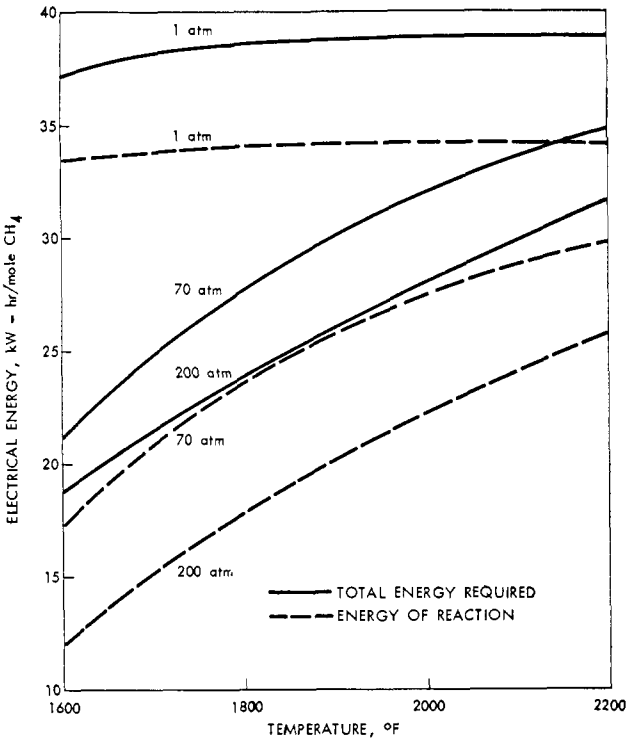


**Figure 10. Gasifier energy required to produce methane based on 90% carbon conversion and equilibrium steam conversion**

would be purified, shift converted, and methanated. The series of steps would be similar to those shown in Figure 8 except that only a single stage each of shift conversion and carbon dioxide absorption would be needed. Although the alternate process would be simpler because of the absence of the hydrogasifier, it would not be as thermally efficient. Estimated energy requirements for the gasifier are presented in Figure 10 for 90% carbon conversion and equilibrium steam conversions. The total energy requirement is based on the method of heat recovery illustrated by Figure 2. For smaller carbon conversions, the energy needed would be greater. For example, at 200 atm, the total gasifier energy requirement is 9 to 14% greater over the indicated temperature range for a carbon conversion of 50% than for a carbon conversion of 90%. To minimize energy consumption, the gasifier should be operated at a high pressure and as low a temperature as possible. Consequently the lower temperature limit would be determined by the kinetics of the gasification reactions. The theoretical yields of methane for the entire process can be determined by using the expressions

$$\text{Yield on carbon} = 0.50\ X \tag{16}$$

$$\text{Yield on steam} = 0.5\ (x_1 + x_3) \tag{17}$$

These expressions indicate that the yield on carbon would not be affected by gasifier temperature or pressure, whereas the yield on steam would be.

The production of methane from char (pure carbon) requires more electrical energy than the integrated hydrogasification process being developed by the Institute of Gas Technology, since this process utilizes coal containing a significant amount of hydrogen. Moreover, the first stage of the hydrogasifier operates at a temperature below 1600°F which improves the equilibrium conversion to methane.

## Present Status and the Future

The chemistry involved in gasifying carbon with steam is well-known, and many of the physiochemical parameters of this reaction system have been determined. On the other hand, the electrofluid bed system requires extensive development even though a number of bench-scale units have been tested and at least two low-pressure commercial units have been used for producing hydrogen cyanide (Shine, 1971). Economical large-scale applications under high pressures require the development of long-life electrodes and efficient power controls. In this regard silicon carbide and stainless steel electrodes appear promising for gasifying carbon with steam. A better understanding of the electrical characteristics is needed to properly design large units. Fortunately research in these areas is continuing and future development of the electrofluid system seems assured.

## Nomenclature

$D$ = diameter of gasifier, dimensionless
$F$ = feed ratio, mol $H_2O$/mol C
$\Delta H_i$ = standard heat of reaction for the $i$th reaction, Btu/lb mol $H_2O$ converted
$\Delta H_r$ = overall heat of reaction, Btu/lb mol $H_2O$ fed
$p$ = pressure, atm
$Q_e$ = electrical energy supplied to gasifier, Btu/lb mol C fed
$Q_s$ = heat energy transferred in steam superheater, Btu/lb mol C fed
$t$ = temperature, °F
$X$ = total carbon conversion, mol C converted/mol C fed
$x_1$ = steam converted by Reaction 1, mol $H_2O$ converted/mol $H_2O$ fed
$x_2$ = steam converted by Reaction 2, mol $H_2O$ converted/mol $H_2O$ fed
$x_3$ = carbon converted by Reaction 3, mol C converted/mol $H_2O$ fed

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